

# An alternate theoretical approach to diffusion bonding

Miguel Lagos\*

*Facultad de Ingeniería, Universidad de Talca, Campus Los Niches, Curicó, Chile*

César Retamal†

*Facultad de Ingeniería, Universidad de Talca, Campus Los Niches, Curicó, Chile and*

*Université de Paris Ouest Nanterre La Défense,*

*Laboratoire de Thermique Interfaces Environnement (LTIE),*

*EA4415, GTE, 50 rue de Sèvres, 92410 Ville d'Avray, France*

(Dated: June 15, 2010)

On the basis of a previous theoretical approach to the plastic flow of highly refined materials, a physical explanation for diffusion bonding is essayed, which yields closed-form equations relating the bonding progress with time, temperature, applied pressure and the constants characterizing the material. Excellent agreement with experiment is attained, with no adjustable parameter. In the novel scheme, diffusion bonding is caused by the interpenetration of the two surfaces at the grain level. The process is driven by the strong tensile stress field induced in the plane of the interface by the plastic deformation in the normal direction. The grain boundaries of each joining surface yield to host grains of the other surface, releasing this way the internally generated tensile stresses. Voids gradually close with the increment of the interpenetrated areas. In this scheme bonding is not a matter of contacting and atomic interdiffusion, but of grain exchange.

PACS numbers: 62.20.fq, 62.20.fk, 62.20.mq, 83.50.Uv

Diffusion bonding (DB) is a solid state high temperature process for making a monolithic joint through matter transportation across the interface between the materials being joined, with no melting at the bond line. It is a costly time-consuming technique for producing top quality bonds, so that neither metallurgical discontinuities nor voids can be detected all over the former interface. Though not worthy for mass production assembly lines yet, millions people are benefited every day by diffusion bonded artifacts. The titanium hollow blades, their internal reinforcing honeycomb structure, and the central disc of the wide chord fan of a present day turbofan commercial aeroengine, which generates about 80% of the total engine thrust at take-off, integrate a single component diffusion bonded unit. On the basis of a recently published theoretical approach to the plastic flow of highly refined materials, we essay here a physical explanation for DB and derive a closed-form equation relating bonding time, temperature, applied pressure and the constants characterising the material, which exhibits excellent agreement with available experimental data, and with no adjustable parameter. In our theoretical approach, DB has little connection, if any, with atomic diffusion.

For two decades, DB combined with superplastic forming (SPF) has been a standard technique for producing aircraft structural components and engine parts, particularly because SPF yields practically no residual stress and neither spring back from die after releasing the forming pressure. Besides, DB eliminates fasteners, yielding

stronger more reliable unions, saving weight and allowing better design [1, 2]. However, the suitability of the process for mass production is yet questionable, mainly by the long bonding times involved and because DB does work just for a short list of known materials. The precise knowledge of the microscopic mechanisms taking part in the process, leading to the physical equations governing its evolution, would be of great value as scientific knowledge, as well as a tool for the search of new improved materials and process optimisation.

The theoretical literature on the subject assumes that any pair of clean metallic surfaces will bond if they are brought together within the range of interatomic forces. Bonding happens by atomic interdiffusion across the interfaces at the contacting surface sectors, which initially are isolated and scarce by the surface roughness. Contacting area gradually increases by plastic deformation and atomic interdiffusion. The large differences in bonding ability observed for the various alloys are ascribed to the chemical and mechanical stability of their surface oxide layers, which may obstruct atomic exchange [1, 3, 4].

In our scheme the process is quite different. It takes place mainly by the active migration of grains that cross the joint line in the two senses, rather than intergrain diffusion of individual atoms. Grain exchange is much more efficient and demands surface proximity of the order of just the grain size, instead of the much smaller atomic distances. The driving force for the interfacial grain exchange arises from the internal stress which plastic deformation necessarily induces in the plane normal to the flow direction.

DB has been observed in highly refined materials constituted by small equiaxed grains, whose plastic deformation conserves the grain mean size  $d$  and overall shapes.

\*Electronic address: mlagos@utalca.cl

†Electronic address: ceretamal@utalca.cl

Thus, grains slide past each other over relatively long distances, accommodating their shapes to preserve matter continuity, with grain boundaries maintaining mechanical integrity and coherence. Hence grains evolve by the action of two kinds of forces that may be of very different strength: those making their boundaries to slide and the induced local forces responsible for the continuous grain reshaping. The stronger one will determine the plastic flow rate.

Recent papers show that excellent agreement with experiment is attained from ascribing the rate control to the intergrain forces that make adjacent grains to slide [5–8]. These forces have been examined by theory [5, 6], Montecarlo computer simulation [11], experiment [10] and molecular dynamics simulation [9]. Said studies clearly show that the onset of grain boundary sliding is associated to a boundary mechanical instability which demands that the shear stress in the plane of the common boundary of the sliding grains be higher than a critical value  $\tau_c$ . Then the grains start sliding with a relative velocity  $\Delta \vec{v}$  which may be proportional to: (a) The difference between the in-plane overcritical shear stress and the threshold stress  $\tau_c$ . (b) The bare in-plane shear stress, and  $\Delta \vec{v}$  jumps from zero to the proper value when  $\tau_c$  is surpassed. We refer to these two options as force models A and B, respectively [8].

As the two models can be fully worked out without additional assumptions, comparison with experiment determines what model is the right one for a specific system. It has been shown that model A works for a series of aluminium and titanium alloys [6, 7], fitting the experimental data within the experimental uncertainties, and model B gives very good results for the steel Avesta 2304 [8].

The technical details for deriving the plastic flow of the polycrystalline medium from the hypotheses stated above are in the literature [5–8]. Summarising, given the stress tensor  $(\sigma_{ij})$ ,  $i, j = x, y, z$ , the force law A or B determines the relative velocities  $\Delta \vec{v}(\theta, \phi)$  between adjacent grains. Angles  $\theta$  and  $\phi$  characterise the grain boundary orientation. One can readily realise that

$$\frac{\partial v_i}{\partial x_j} = \frac{1}{d} \langle \Delta v_i \rangle_j, \quad (1)$$

where  $\vec{v}$  is the velocity field of the material medium and symbol  $\langle \dots \rangle_j$  stands for the average over all boundary orientations along the  $x_j$  axis in the positive sense. (Notice that we denote either  $i, j = x, y, z$  or  $x_i = x, y, z$ ,  $i = 1, 2, 3$ ). Making the averages in explicit way, closed-form equations relating  $\partial v_i / \partial x_j$  with the stresses are derived from Eq. (1). Rotation invariants like  $\nabla \cdot \vec{v}$  and  $\nabla \times \vec{v}$ , or the components  $\dot{\epsilon}_{ij}$  of the strain rate tensor, which is the symmetric part of tensor (1), can be constructed with them.

For uniaxial external stress  $\sigma$  along the  $z$  axis and cylindrical symmetry with respect to the same axis, so

that  $\sigma_{xx} = \sigma_{yy} \equiv \sigma_\perp$  and  $\sigma_{ij} = 0$  for  $i \neq j$ , the strain rate along  $z$  is shown to be

$$\dot{\epsilon} = \frac{\partial v_z}{\partial z} = s \frac{\tau_c Q(p)}{2d} \left[ \cot(2\theta_c) + \alpha \left( 2\theta_c - \frac{\pi}{2} \right) \right], \quad (2)$$

and

$$\begin{aligned} \nabla \cdot \vec{v} = & -s \frac{\tau_c Q(p)}{2d} \left[ \frac{1 - \cos(2\theta_c)}{\sin(2\theta_c)} - 2\theta_c \left( \alpha + \frac{2}{\pi \sin(2\theta_c)} \right) \right. \\ & \left. + \frac{2}{\pi} (1 - 2\alpha) \cos(2\theta_c) + \alpha \frac{\pi}{2} \right], \end{aligned} \quad (3)$$

where  $s$  is the sign of the deviatoric stress  $\sigma + p$ , being  $p$  the pressure  $p = -(\sigma + 2\sigma_\perp)/3$ , and the coefficient  $Q(p)$  is the proportionality factor between the relative speed of adjacent grains and the resolved overcritical shear stress. Temperature  $T$  apart,  $Q(p)$  depends only on the pressure invariant  $p$  because it must keep unchanged for any boundary orientation. The auxiliary variable  $\theta_c$  is defined by

$$\sin(2\theta_c) = \frac{4\tau_c}{3|\sigma + p|} \quad (4)$$

and  $\alpha$  takes the values 1 or 0 for force models A or B, respectively.

Eq. (3) discloses a remarkable consequence of the existence of a finite threshold stress for grain sliding. As  $\nabla \cdot \vec{v} = \dot{V}/V \neq 0$ , where  $\dot{V}/V$  is the dilation rate per unit volume, the specific volume is not conserved in plastic flow if  $\tau_c \neq 0$ . As grain volume variations can only be elastic, one can recall Hooke's law and write  $\nabla \cdot \vec{v} = -(1/B)\dot{p}$ , where  $B$  is the bulk elastic modulus. Combining this with equation (3) it follows the equation

$$\begin{aligned} \dot{p} = & s \frac{B\tau_c Q(p)}{2d} \left[ \frac{1 - \cos(2\theta_c)}{\sin(2\theta_c)} - 2\theta_c \left( \alpha + \frac{2}{\pi \sin(2\theta_c)} \right) \right. \\ & \left. + \frac{2}{\pi} (1 - 2\alpha) \cos(2\theta_c) + \alpha \frac{\pi}{2} \right] \end{aligned} \quad (5)$$

for the evolution of  $p$  on plastic flow.

Pressure  $p$  monotonically increases or decreases for plastic axial stretching ( $s > 0$ ) or shrinking ( $s < 0$ ), respectively. As  $\sigma$  is the externally applied stress, the increase of  $|p|$  must be attributed to the variation of

$$\sigma_\perp = -\frac{1}{2}(\sigma + 3p), \quad (6)$$

which represent an internal radial stress field normal to the plastic flow direction. Settling  $\sigma_\perp = 0$  (or  $p = -\sigma/3$ ) at start, when the strain  $\epsilon = 0$ , is a natural initial condition. However, as the deformation proceeds,  $\sigma_\perp$  becomes finite, taking negative values on stretching, and

hence contributing to neck formation, and positive ones on axial compression. Both  $p$  and  $\sigma_{\perp}$  grow at a high rate because  $B \sim 10^5$  MPa. If the plastic flow is interrupted, the deformation induced stress field  $\sigma_{\perp}$  will remain as a residual stress.

We must remark that the equations written above assume cylindrical symmetry. In strict rigor, they hold for an axially symmetric sample or at any point of an infinite medium. Symmetry breaking surfaces make the problem much more complex. However, there is a practical situation for which Eqs. (2) and (5) hold strictly: if the material is finite in the  $z$  direction, but infinite in the  $xy$  plane, one can anyway assume cylindrical symmetry at any point of it. Hence, Eqs. (2) and (5) are obeyed by an extended flat plate subjected to a compressive stress  $\sigma$  normal to its plane, and strong enough to produce plastic deformation. In such case the material has no means to relax the tensile stress  $\sigma_{\perp}$ , which progressively builds up in the  $xy$  plane upon plastic compression in the  $z$  direction.

However, there is again a practical situation for which the extended plate is able to relax the induced tensile stress, at least partially, at the surfaces. If we compress together two plates with surface irregularities of the order of the grain size  $d$ , the grains of one of them may cede to  $\sigma_{\perp}$  and open up if the consequent interstice is simultaneously filled by one or more grains provided by the other surface. The process is reciprocal, and the interpenetration of the two surfaces at the grain level provides a mechanism to release the tensile cumulative stress  $\sigma_{\perp}$ . The interpenetration of the contacting surfaces makes their areas to increase, filling this way the voids left by the surface irregularities.

The rate of bonding is dictated by the growing rate of the total interpenetrating surface area  $A$ . A good criterion is to assume that  $A$  varies at a speed that keeps  $\sigma_{\perp} = 0$ . We have that  $\dot{A}/A = \nabla \cdot \vec{v} - \partial v_z / \partial z$ . Hence  $\dot{A}/A$  is obtained subtracting Eq. (2) from Eq. (3), with  $s = -1$  and  $p = -\sigma/3$ , or, equivalently,  $\sin(2\theta_c) = 2\tau_c/|\sigma|$ . Additionally, the coefficient  $Q(p)$  was studied in Refs. 5 and 6. The mechanisms for grain boundary sliding considered there give

$$\frac{Q(p)}{4d} = C_0 \frac{\Omega^*}{k_B T} \exp\left(-\frac{\epsilon_0 + \Omega^* p}{k_B T}\right), \quad (7)$$

where  $k_B$  is the Boltzmann constant, the coefficient  $C_0$  depends only on the grain size  $d$ , the constant  $\epsilon_0$  is the energy necessary for evaporating a crystal vacancy from the grain boundary, and  $\Omega^*$  measures the sensitivity of this energy to stress. Combining all this, one has that

$$\begin{aligned} \lambda = \left(\frac{\dot{A}}{A}\right)_0 &= 2C_0 \frac{\Omega^* \tau_c}{k_B T} \left[ \frac{|\sigma|}{2\tau_c} - \frac{2}{\pi} \frac{|\sigma|}{2\tau_c} \arcsin\left(\frac{2\tau_c}{|\sigma|}\right) \right. \\ &\quad \left. + \frac{2}{\pi} (1 - 2\alpha) \sqrt{1 - \left(\frac{2\tau_c}{|\sigma|}\right)^2} \right] \\ &\times \exp\left(-\frac{\epsilon_0 + \Omega^* |\sigma|/3}{k_B T}\right) \quad (\sigma_{\perp} = 0), \end{aligned} \quad (8)$$

where the subscript of  $(\dot{A}/A)_0$  indicates that equation (8) gives the initial grow rate of the bonded area fraction, when  $\sigma_{\perp} = 0$ .

The condition  $\sigma_{\perp} = 0$  will cease only once voids be almost filled. Hence, what matters in practice is the initial binding rate  $\lambda$ , and it can be assumed that  $\dot{A}$  vanishes when  $\sigma_{\perp}$  becomes finite because  $A$  has no free room to grow. On this basis one can write the equation

$$\dot{A} = \lambda \frac{A_1}{A_0} (A_1 - A), \quad (9)$$

where  $A_1$  is the total area to be bound,  $A_0$  is the initial contact area, constituted by  $A_1/A_0$  small islands which are assumed already joined.  $A$  is the bonded area at time  $t$  and  $A_1 - A$  the unbound area. The coefficient  $\lambda$  measures the intrinsic ability of the material for making DB joints. Eq. (9) has the solution

$$\frac{A}{A_1} = 1 - \exp\left(-\lambda \frac{A_1}{A_0} t\right), \quad (10)$$

which gives the time dependent area fraction bonded.

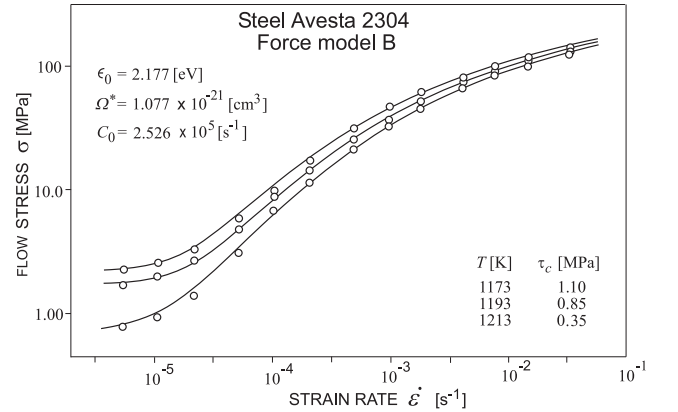


FIG. 1: Flow stress *versus* strain rate for the steel Avesta 2304 at  $T = 1173, 1193$  and  $1213$  K. Circles represent experimental data taken from Ref. 12 and solid lines depict Eq. (2) with  $\alpha = 0$  and the constants shown in the insets. The parameter  $\tau_c$  exhibits some temperature dependence.

Pilling, Wang and Ridley [12] authored one of the very scarce experimental observations of the time progression of DB available in the literature. Together with the time

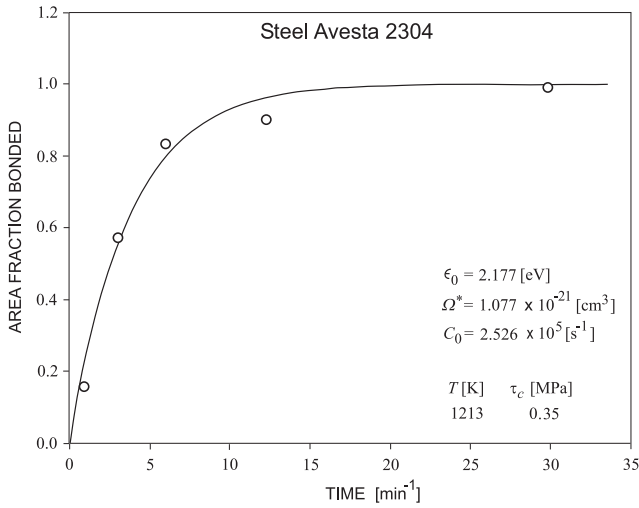


FIG. 2: The variation of the area fraction bonded with time for steel Avesta 2304 at  $T = 1213$  K and  $\sigma = 3.0$  MPa. Circles represent experimental data taken from Ref. 12. The solid line represents equation (10) with  $\lambda$  given by Eq. (8) and the material constants obtained from the fit shown in Fig. 1. The theoretical curve has no adjustable parameter.

evolution  $A(t)/A_1$  of two bonding surfaces of the steel Avesta 2304 at  $T = 1213$  K and  $\sigma = 3.0$  MPa, they reported also the flow stress  $\sigma$  for plastic stretching versus strain rate  $\dot{\epsilon}$  at several temperatures. Therefore, their data allows us to test Eq. (2), and Eqs. (8) and (10) independently. Fig. 1 shows the fit given by Eq. (2) to the

flow stress data ( $\epsilon \approx 0$ ) at three temperatures, which gives us precise values for the constants  $\epsilon_0$ ,  $C_0$ ,  $\Omega^*$  and  $\tau_c$  of the material.

Inserting these material constants in Eq. (8), together with  $\sigma = 3.0$  MPa and  $T = 1213$  K, it is obtained that  $\lambda = 4.11 \times 10^{-5} \text{ s}^{-1} = 2.47 \times 10^{-3} \text{ min}^{-1}$ . On the other hand, Pilling *et al.* [12] reported that the two joining steel surfaces in the DB experiment initially display very small contact sectors, of less than  $4 \mu\text{m}$ , distant  $l = 50 - 55 \mu\text{m}$  between them. We take for the ratio  $A_1/A_0 = (l/d)^2$  because the size of the initial bound spots cannot be smaller than the grain size  $d = 5 \mu\text{m}$ . Therefore, taking  $l = 52 \mu\text{m}$  it turns out  $A_1/A_0 = 108.16$ . The solid line in Fig. 2 depicts Eq. (10) after replacing in it all these constants, together with the experimental data of Pilling *et al.* The agreement is quite impressive, particularly because it was attained with no adjustable parameter.

In summary, we obtain close agreement with experiment from interpreting DB as the interpenetration of the two surfaces at the grain level. The process is driven by the strong tensile stress field induced in the plane of the interface by the plastic deformation in the normal direction. At each point of contact the grain boundaries of one surface yield to host grains of the other surface, and reciprocally, releasing this way the internally generated tensile stresses. Voids close by the gradual increment of the interpenetrated contacting areas. Hence, bonding is not a matter of contacting and atomic interdiffusion, but of grain exchange.

- 
- [1] L. D. Hefti, *J. Mat. Eng. Perform.* **17**, 178 (2008).
  - [2] T. G. Nieh, J. Wadsworth and O. D. Sherby, **Superplasticity in metals and ceramics** (Cambridge, UK 1997).
  - [3] J. Pilling, *Mater. Sci. Eng.* **100**, 137 (1988).
  - [4] N. Ridley, M. T. Salehi and J. Pilling, *Mater. Sci. Technol.* **8**, 791 (1992).
  - [5] M. Lagos, *Phys. Rev. Lett.* **85**, 2332 (2000).
  - [6] M. Lagos, *Phys. Rev. B* **71**, 224117 (2005).
  - [7] M. Lagos and C. Retamal, *Phys. Scr.* **81**, 055601 (2010).
  - [8] M. Lagos and C. Retamal, submitted to *Phys. Scr.*
  - [9] Y. Qi and P. E. Krajewski, *Acta Mater.* **55**, 1555 (2007).
  - [10] H. Fukutomi, T. Yamamoto, K. Nonomura and K. Takada, *Interface Sci.* **7**, 141 (1999).
  - [11] P. Bellon and R. S. Averback, *Phys. Rev. Lett.* **74**, 1819 (1995).
  - [12] J. Pilling, Z. C. Wang and N. Ridley, in *Superplasticity and Superplastic Forming 1998*, p. 297, ed. by A. K. Ghosh and T. R. Bieler (The Minerals, Metals and Materials Society, Warrendale, PA, 1998).